



On the Importance of Reliable Background Concentrations of Ozone for Regional Scale Photochemical Modelling

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Abstract. Ozone throughout the troposphere is subject of significant temporal and spatial variability due to photochemical production in the planetary boundary layer and free troposphere, stratospheric intrusions, convective events and long range transport. However, high resolving observations of ozone in the troposphere are generally rare today. That is of special disadvantage for limited area models, which represent mathematically a differential equation system with an initial and boundary problem. As ozone concentrations usually increase from the earth surface to the stratosphere, a proper choice of the background ozone concentrations is necessary to reproduce or even predict the amount and distribution of ozone in a specific region of interest.

In this paper the impact of background concentrations of ozone on regional scale model results is analysed during a summer smog episode over Europe. For this purpose ozone is artificially partitioned into individual categories. For each category, transport and chemical transformation is calculated separately. Initial and boundary concentrations of ozone dominate total ozone concentrations increasingly with height. But also in the planetary boundary layer they contribute with more than 30% to the total ozone changes and are therefore far from being negligible. Moderately modified assumptions of background ozone concentrations reveal an uncertainty of near surface ozone concentrations of 5–15% depending on the weather situation.

Key words: background concentrations, ozone, regional modelling, vertical mixing.

1. Introduction

Elevated concentrations of ozone in the lower troposphere during summertime are still a major air pollution issue in the industrialised areas of the world. High near surface ozone concentrations damage human health, vegetation and materials. In the planetary boundary layer (PBL) ozone is a secondary pollutant formed by a series of photochemical reactions. Among its precursor substances are nitrogen oxides and volatile organic compounds (VOC). During the last decades national environmental agencies have developed and applied emission control and reduction strategies for these precursor substances to reduce near surface ozone concentrations. As a consequence peak ozone concentrations have decreased during the last years (Umweltbundesamt, 2000), however, the background ozone level

has increased (Umweltbundesamt, 2000; Crutzen, 1995). As ozone may exist for several days in the lower troposphere, long-range transport is possible which complicates the understanding of measured ozone concentrations. For the interpretation of observed ozone concentrations and for the estimate of the effect of emission control strategies, atmosphere-chemistry models are helpful tools. A huge number of regional scale air pollution and dispersion models with different degrees of complexity have been developed and applied in recent years. Besides hindcast experiments to improve the present understanding of tropospheric photochemistry, they have also been further developed to predict surface ozone concentrations.

The comparison of measured and modelled ozone concentrations in the PBL reveals that up to now models cannot reproduce or predict daily maximum ozone concentrations with an accuracy of more than about 10 to 20% (Schaller and Wenzel, 2000). A number of uncertainties are responsible for this disagreement. One of the most crucial uncertainties is attributed to the emission data (Placet *et al.*, 2000). The collection of the spatially and temporally varying fluxes of emitted species from anthropogenic activities is a difficult task with an estimated uncertainty of more than 50% for high resolution traffic and solvent emissions (Kühlwein *et al.*, 1999; Hassel *et al.*, 1995). Biogenic emission fluxes of e.g. isoprene are even more uncertain (Andronache *et al.*, 1994). Another important topic is the determination of the physical state of the atmosphere in space and time. Although the large scale weather situation can be simulated with high confidence for weather forecast or hindcast experiments, the simulated values at a single grid point might differ considerably from reality. The uncertainties in physical parameterisations of meteorological models (subgrid scale transport like turbulence and moist convection, soil processes, radiative transfer to mention a few of them) and the horizontal and vertical model resolution are responsible for these differences. Errors or uncertainties of the simulated meteorological variables directly influence the calculation of transport and transformation processes of chemical species. The chemical mechanism of the models which is chosen to determine gas phase (Gao *et al.*, 1996), aqueous phase and perhaps also aerosol phase chemical transformations is another cause of uncertainty.

A lot of work is on the way to further improve the above mentioned issues. However, one additional important topic that has been often ignored so far is the influence and uncertainty of initial and boundary concentrations of long living species like ozone and carbon monoxide on regional model results. In contrast to most of the primarily emitted and secondarily formed pollutants from anthropogenic activity, ozone concentrations increase from the earth surface to the stratosphere with a maximum at about 20 to 25 km height. In the free troposphere and the stratosphere, the chemical lifetime of ozone is significantly longer than in the PBL. However, strong day to day ozone variability on the scale of synoptic meteorological disturbances has been observed by ozonesonde measurements throughout the troposphere during spring and summer in the remote area of the North Atlantic (Oltsman *et al.*, 1996) as well as over the continental areas of

Europe (regular ozonesonde observations at several stations, for example Lindenberg and Hohenpeissenberg in Germany). Due to the vertical gradient, long range transport, stratospheric intrusions, convective events, photochemical production in the PBL and free troposphere, vertical ozone profiles vary significantly in time and space. Recently, the importance of intercontinental transport of photooxidants and precursor substances in northern mid-latitudes has been emphasised again after early indications by Parish *et al.* (1993). The authors speculated that transport from North American sources may play a role in the increase of ozone over Europe, particularly in the free troposphere. This assumption was based on measurements of transport and transformation of anthropogenic emissions from industry and traffic released into the PBL at the east coast of the United States of America. However, during summer North American forest fire emissions are an additional important source for photooxidants and the intercontinental transport (Forster *et al.*, 2001). Stratospheric intrusions of ozone rich air masses due to strong convective events or tropopause foldings also contribute to the long range transport and elevated ozone levels in the troposphere, sometimes far away from the region of origin (Stohl and Trickl, 1999). In occasionally occurring thunderstorms NO production by lightning is another source for photochemical ozone production in the free troposphere, especially during summer. If the goal of a limited area model study is to reproduce or even predict peak ozone concentrations in the PBL, it is important to quantify the uncertainty caused by the high ozone background in the troposphere as well as the uncertainty caused by the above discussed spatial and temporal variability.

One former article investigating systematically the sensitivity of atmospheric concentrations as simulated by a regional chemical model (Brost, 1988) concludes that:

- as the simulation proceeds, initial concentrations decline in importance,
- the effect of boundary concentrations normally increase with height,
- the longer the chemical lifetime of the species, the more important are the initial and boundary concentrations.

Brost (1988) discussed results for a single specific 3-day episode in April 1981. During this episode, a cyclonic storm developed and moved across the north eastern United States. These results may not be generalised to other regions and seasons. Especially during a summer air pollution episode with a stagnant high pressure system, the importance of initial and boundary concentrations might differ. In summer and spring there is substantial vertical transport by clouds between the PBL and the free troposphere. However, the lack of rapid horizontal transport aloft during summer means that the initial conditions remain important for a longer period of time while the lateral boundary conditions may be less important. As ozone forecast is desired during summer smog episodes when peak ozone concentrations occur, a detailed analysis of the influence of ozone initial and boundary concentrations on regional scale model results will be presented in this paper for a summer smog episode over Europe. The model description and set up is introduced in Section 2.

In Section 3 the model results will be described and analysed. Section 4 draws conclusions and gives an outlook.

2. Model Description and Set Up

The online atmosphere-chemistry model REMO (Langmann, 2000), which is used in the present study, determines for every timestep of the model meteorology, transport and photochemistry. The dynamical part and the physical parameterisation routines are taken from the regional weather forecast model system EM/DM of the German Weather Service (Majewski, 1991). The model can also use the physical parameterisations of the global ECHAM-4 model (Roeckner *et al.*, 1996), but this configuration was not used for the present investigation. Tracer transport is calculated as horizontal and vertical advection according to the algorithm of Smolarkiewicz (1983), as convective up- and downdraft by a modified scheme of Tiedtke (1989), as vertical diffusion (Mellor and Yamada, 1974) and as dry deposition (Wesley, 1989). The RADM II chemical scheme (Stockwell *et al.*, 1990) describes production and loss by chemical reactions in the gas phase. Aqueous phase chemistry processes and wet removal are implemented according to Walcek and Taylor (1986). Photolysis rates are calculated as in Madronich (1987) and Chang *et al.* (1987). Anthropogenic emission data are provided by the Institute of Energy Economics and the Rational Use of Energy, Stuttgart, Germany (B. Wickert, personal communication) and are therefore different from the study of Langmann (2000). Biogenic VOC emissions are determined based on Guenther *et al.* (1991, 1993).

Currently, the initialisation of regional air pollution models for a certain episodic simulation cannot be entirely based on observed concentrations throughout the troposphere because of a lack of observations. In most cases, only near surface measurements are available which, in addition, are restricted to only a few key chemical species. Hence, the REMO model initialisation for chemical trace species follows the approach proposed by Chang *et al.* (1987). From available measurements during recent years vertical profiles for each model species are derived. A model simulation is started with these vertical profiles horizontally distributed over the entire model domain and with realistic meteorology and emissions. The only exception is H_2O_2 which is initialised and updated at the lateral boundaries via a dew point correlation (Daum *et al.*, 1990). All other species concentrations are fixed at the lateral boundary grid points – that is a ring of the dimension of one grid cell around the model domain – throughout the simulation because the information about their spatial and temporal variability during a given episode is poor. In order to minimise the influence of the boundaries upon the inner model domain under inflow conditions, the lateral boundaries should therefore be located relatively far away from the region of interest. A detailed description of the REMO model with coupled chemistry is presented by Langmann (2000).

In the current study the REMO model is applied with 20 vertical layers of unequal thickness between the earth surface and the 10 hPa pressure level. A terrain following hybrid pressure-sigma coordinate is used in the vertical. The horizontal resolution is 0.5° on a spherical rotated grid. The model domain of 1.17×10^7 km² covers Europe, as shown in Figure 1. The prognostic equations for surface pressure, temperature, specific humidity, cloud water, horizontal wind components and chemical trace species concentrations are written on an Arakawa-C grid. The model is run in the forecast mode with respect to the model physics. 6 hour meteorological analysis data provided by the German Weather Service are used as initial and lateral boundary information. Starting at 0 UTC every day a 30 hour forecast is computed. The first six hours of the consecutive meteorological only forecasts are neglected to account for a spin up time. During the following 24 hours atmospheric physics and chemistry are calculated continuously. At 6 UTC on the following day a discontinuity in the physical state of the model atmosphere is introduced due to the new daily meteorological forecast, whereas tropospheric chemistry is calculated continuously. It is a usual procedure to start a model every day with meteorological analysis data because this way the model atmosphere is forced to stay close to the real weather situation.

To assess the influence and uncertainty of initial and boundary conditions on calculated ozone concentrations, ozone is artificially partitioned into seven individual categories as summarised in Table I. For a better understanding of this split, the spatial distribution of each ozone category at the initialisation time step is illustrated schematically in Figure 2. As photochemically produced ozone, $O_{3,pr}$, is set to zero when initialising the model, it does not appear in Figure 2. The separation into stratospheric ozone, $O_{3,s}$, and tropospheric ozone (categories 4–7) represents only a coarse approximation because it neglects the variation in the altitude of the tropopause. Each category is transported separately within the model domain. Thus, after the initial time step, each individual ozone category is allowed to exist anywhere inside the model domain. The total ozone concentration, $O_{3,t}$, at a single grid point is always equal to the sum of the seven ozone categories (see Table I for the definition of the symbols):

$$O_{3,t} = O_{3,i} + O_{3,pr} + O_{3,s} + O_{3,bn} + O_{3,be} + O_{3,bs} + O_{3,bw} . \quad (1)$$

As discussed before, the concentrations at the lateral boundary grid points are held constant and equal to the initial conditions (see Figure 2) throughout the entire model simulation. Inside the model area, the concentrations of each ozone category might decrease due to chemical loss reactions, dry deposition at the earth surface, and transport to outside the model area. Photochemical ozone production leads to an increase of concentration of $O_{3,pr}$ whereas the only source of ozone of categories 3 to 7 is transport across the lateral model boundaries into the model domain.

Similar approaches with tagged species have been applied in global model studies. In Lamarque *et al.* (1996) a three-dimensional global model simulation is described which intends to investigate the relative importance of the different

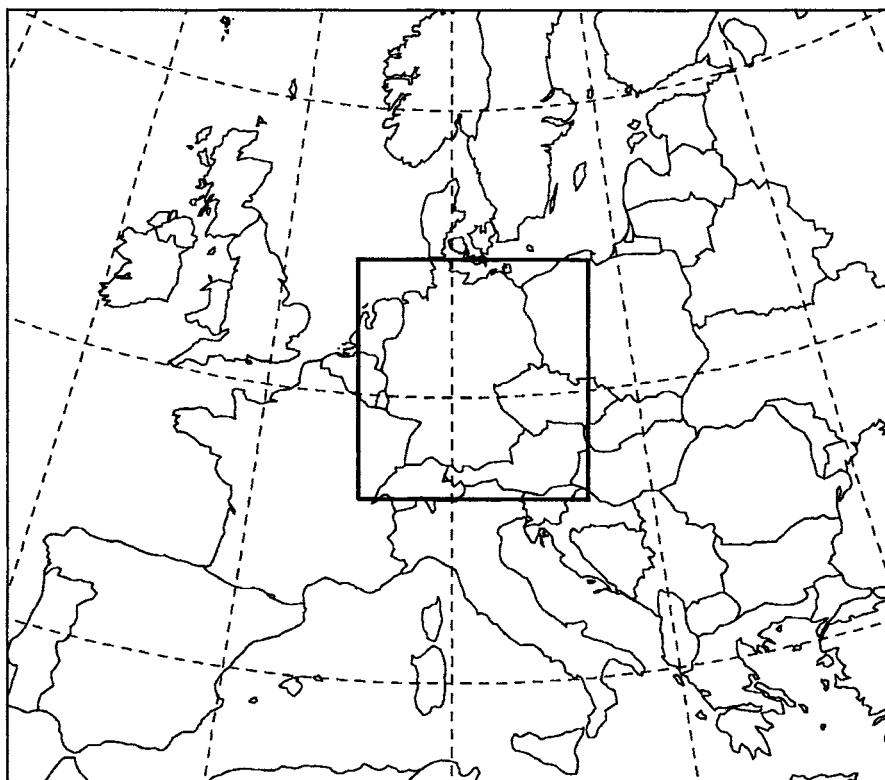
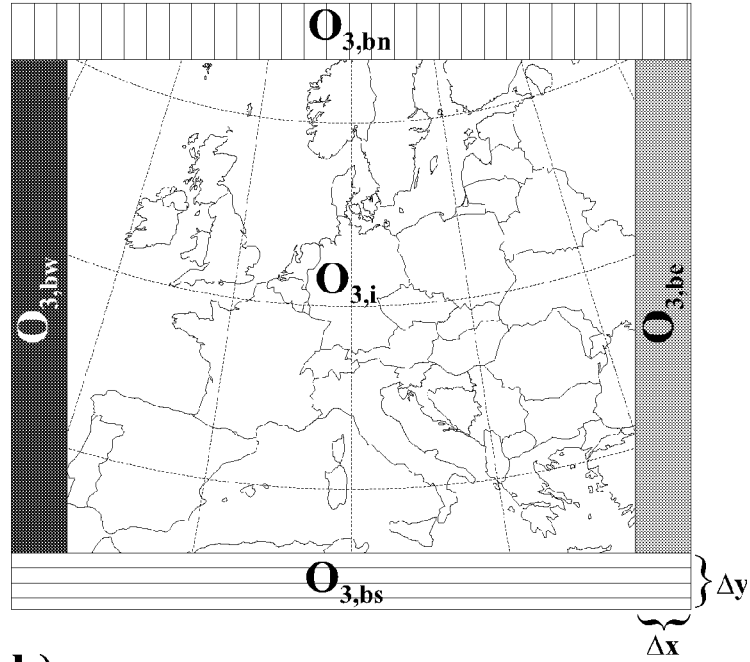


Figure 1. Total model domain and German subarea.

Table 1. Definition of ozone categories

	O ₃ category	Definition
1	O ₃ , initial	Initial O ₃ distribution inside the model domain
2	O ₃ , pr roduced	Photochemically produced O ₃ inside the model domain
3	O ₃ , stratosphere	O ₃ originating from the last lateral boundary grid points above 10 km height
4	O ₃ , boundary north	O ₃ originating from the last northern lateral boundary grid points up to 10 km height
5	O ₃ , boundary east	O ₃ originating from the last eastern lateral boundary grid points up to 10 km height
6	O ₃ , boundary south	O ₃ originating from the last southern lateral boundary grid points up to 10 km height
7	O ₃ , boundary west	O ₃ originating from the last western lateral boundary grid points up to 10 km height

a)



b)

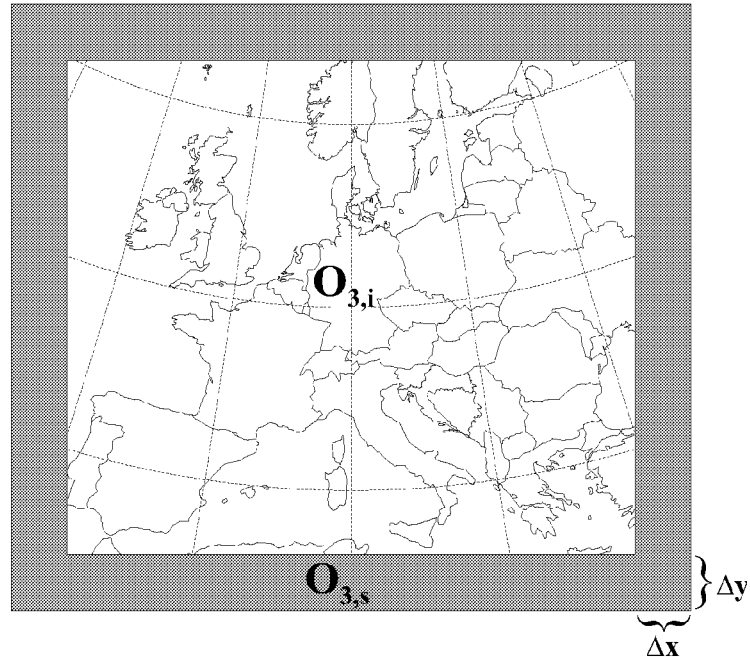


Figure 2. Schematic illustration of the initial distribution of the individual ozone categories (grey shaded) (a) throughout the troposphere up to 10 km height and (b) throughout the stratosphere above 10 km height. The whole box represents the model area. A ring of the dimension of one grid cell (Δx , Δy) around the interior of the model domain is reserved for the boundary concentrations.

odd nitrogen sources in the troposphere by tagging nitrogen atoms according to their sources. The relative contribution of individual sulfur sources to the global atmospheric sulfur burden is investigated by Graf *et al.* (1997) by calculating not only the transport of total sulfur dioxide and sulfate but additionally the ratio between the contribution from a specific source to the total sulfur budget. For limited area models a tagged species approach seems somewhat more artificially in comparison to global model applications because of the lateral boundaries and the missing information from sources outside the model domain. Therefore the present study classifies the amount of ozone transported into the domain in several classes of boundary ozone (Table I). These classes represent what has happened outside the REMO model domain. In contrast to Lamarque *et al.* (1996) and Graf *et al.* (1997) who studied N- and S-atoms, respectively, as part of different molecules, we do not intent to follow the individual O-atoms of an ozone molecule through all transport and transformation processes. That would be of desperate concern taking into account the large number of molecules containing O-atoms involved in photochemical reactions. The focus of the present study is the ozone molecule itself. The approach is physically correct. However, the total ozone concentration as determined from Equation (1) is slightly different from the ozone concentration derived when ozone is transported as a single component. Local gradients in the mixing ratios of the individual O_3 categories originating from separating the sources are not existent for unsplitted O_3 . Hence, the non-linear advection scheme of Smolarkiewicz (1983) determines locally different advection. The upstream step of this scheme introduces no differences, but the antidiffusion correction step introduces differences because it involves downstream concentrations. Nevertheless, in the regional average these differences are significantly smaller than 1% and unimportant for the conclusions of this study. The approach is also chemically correct. Each category contributes to the chemical transformations. The sum of all individual transformations equals to the transformation of $O_{3,t}$, so that nothing is neglected in the chemical reaction scheme. This is demonstrated exemplary for the following reaction with k as reaction coefficient:



The amount of ozone lost by reaction (2) is

$$d[O_3]/dt = -k[O_3][NO_2]. \quad (3)$$

Substituting O_3 by the split introduced in Equation (1) allows to separate seven individual differential equations from Equation (3), the sum of all is equivalent to $d[O_3]/t$. Although physically and chemically correct, the presented approach includes some limitations due to the fast photochemical reactions involved. If an ozone molecule is photolysed, it is lost from its category. Even if an ozone molecule is produced immediately afterwards from molecular oxygen and the oxygen radical, it is now categorised as $O_{3,pr}$. Therefore, the presented approach might lead to an overestimation of ozone in category $O_{3,pr}$. That means, this method gives an

estimate of the lower limit of the influence of initial and boundary concentrations of ozone for regional scale modelling.

3. Model Results and Discussion

A summer smog episode over Europe in July 1994 (20–28 July) has been simulated with the atmosphere-chemistry model REMO. The whole episode was characterised by high surface pressure over Central Europe and frequently passing low pressure systems over the British Isles and Scandinavia. Until 25 July dry continental air masses without clouds dominated over Central Europe. On 25 July an occlusion with strongly decreasing intensity entered Germany from the west, accompanied by moist subtropical air and thunderstorms. During the following days the weather situation over Germany remained moist and hot until 29 July, when fresh maritime air was transported towards Germany. A first application of the atmosphere-chemistry model REMO during this episode in July 1994 is described and evaluated against near surface measurements in Langmann (2000). Therefore the current investigation focuses on the interpretation of modelled concentrations of the specific ozone categories introduced in Section 2. First a baseline model simulation will be presented and analysed, then a sensitivity experiment will be discussed.

The temporal development of the individual ozone categories during the summer smog episode is shown in Figure 3. The total ozone mass content in the model domain, $O_{3,t}$, is initialised with 9.8×10^{12} g. Throughout the simulation it is allowed to vary depending on the inflow and outflow conditions, dry deposition and chemical production and loss reactions. The fraction of initialised ozone decreases exponentially with time and becomes less than 10% of $O_{3,t}$ after four days of simulation. After this initialisation period, a steady state situation is reached with variations of less than 10% in the contribution of the individual ozone categories to the total ozone content. The influence of ozone advected into the model domain from the northern, southern and eastern boundary never exceeds 10% whereas ozone imported from western directions contributes to about 20%. Ozone produced photochemically makes up 30% of $O_{3,t}$ with variations reflecting the diurnal cycle. The highest fraction of $O_{3,t}$ (40%) is provided by stratospheric ozone simply because ozone concentrations increase considerably with altitude. However, regional scale air pollution modelling is mainly interested in trace species distributions in the lower troposphere. Stratospheric influences on ozone are therefore of minor interest except when tropopause folding events with ozone-rich intrusions from the stratosphere occur. For the described episode in July 1994 no such a tropopause fold was found by K. Krüger (personal communication).

The next step in the analysis of the impact of initial and boundary concentrations on regional ozone will be carried out for the subarea of Germany (0.74×10^6 km²) as represented in Figure 1. The boundary conditions for this case remain specified at the edge of the large European domain, so that an im-

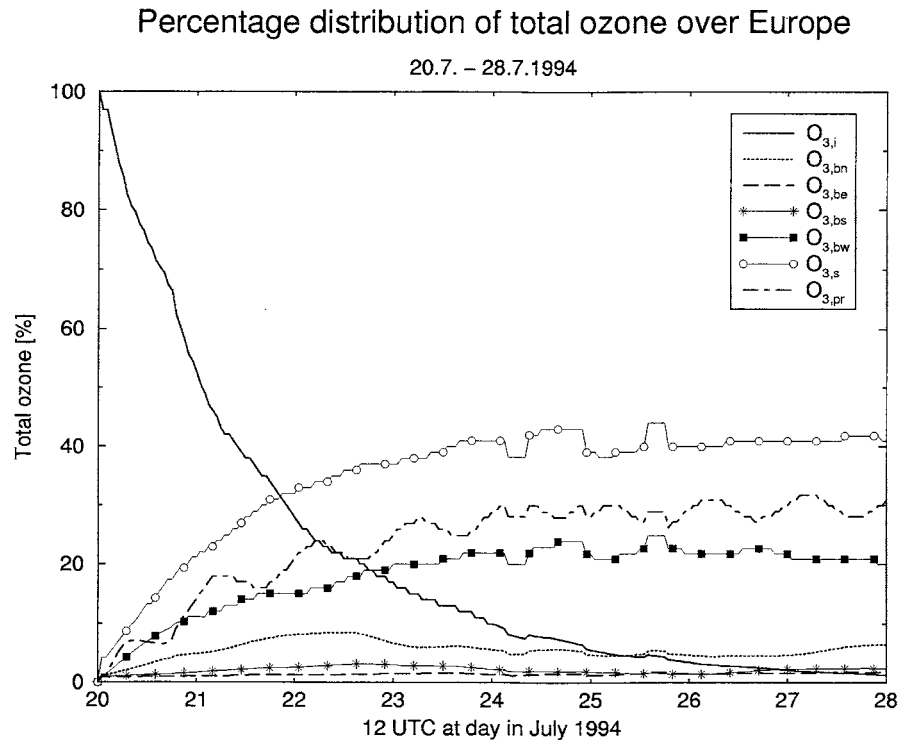


Figure 3. Percentage distribution of total ozone over Europe during the summer smog episode in July 1994.

mediate influence of ozone concentrations from the lateral boundaries is avoided. The temporal development of the total ozone mixing ratio, initial ozone, boundary ozone ($O_{3,b} = O_{3,s} + O_{3,bn} + O_{3,be} + O_{3,bs} + O_{3,bw}$) and photochemically produced ozone in the German subarea is shown in Figure 4 as averaged vertical distribution up to approximately 13 km height. The mean initial ozone distribution over Germany is reduced to less than 10% after five days of the simulation. Between 2 and 6 kilometers altitude, $O_{3,i}$ (due to initial conditions) is the dominant category only during the first days of the simulation. Afterwards ozone in this altitude region of the lower free troposphere consists of $O_{3,b}$ (provided by boundary conditions) and $O_{3,pr}$ (produced by photochemistry) with the $O_{3,pr}$ fraction increasing and the $O_{3,b}$ fraction decreasing towards the earth surface. The contributions of $O_{3,b}$ are mainly determined by stratospheric ozone and by ozone imported from the western boundary. Figure 4 suggests that the initial ozone distribution (except during the first 24 hours of the simulation) and the boundary concentrations of ozone are completely unimportant for the determination of ozone in the PBL, at least during the episode discussed here.

However, as already mentioned in Section 2 the splitting approach might lead to an overestimation of ozone in category $O_{3,pr}$. This statement is supported by

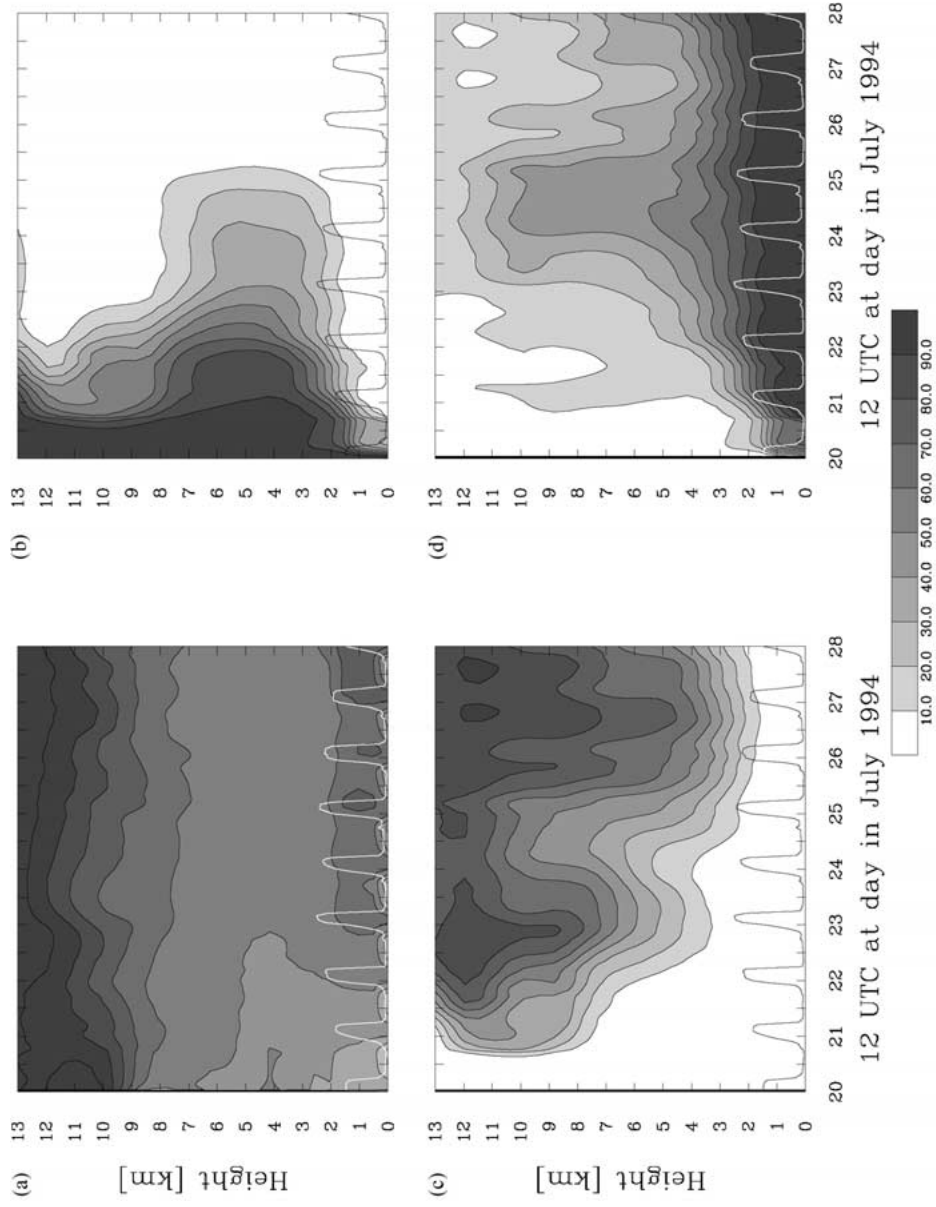


Figure 4. Vertical distribution up to approximately 13 km height of total ozone in ppbv and fraction of initial ozone, boundary ozone and photochemically produced ozone in % as average over Germany and as function of time. The PBL height is indicated in the lower two kilometers. (a) $O_{3,t}$ [ppbv]; (b) $O_{3,i}$ [%]; (c) $O_{3,b}$ [%]; (d) $O_{3,pr}$ [%].

Table II. Percentage contributions of $O_{3,pr}$ and $O_{3,b} + O_{3,i}$ to the total ozone mass changes in the PBL of the German subarea due to horizontal advection (hor. ad.), vertical advection (vert. ad.), vertical diffusion and dry deposition (vert. dif.), aqueous phase chemistry (aq. chem.), moist convection (m. conv.), gas phase chemistry (g. chem.) and as sum over all processes during the eight days episode. The percentage contributions are determined from the absolute values of the respective numbers given in Figure 5.

	$O_{3,pr}$ [%]		$O_{3,b} + O_{3,i}$ [%]	
	Basic run	(Sensitivity run)	Basic run	(Sensitivity run)
hor. ad.	64	(58)	36	(42)
vert. ad.	76	(71)	24	(29)
vert. dif.	100	(100)	0	(0)
aq. chem.	100	(100)	0	(0)
m. conv.	58	(52)	42	(48)
g. chem.	74	(69)	26	(31)
Σ	68	(64)	32	(36)

the analysis of the single processes contributing to the net concentration change of ozone within the PBL over Germany, which provides a different view. Figure 5 presents ozone mass tendencies as grey shaded bars (the black bars indicate results from the sensitivity experiment described below) due to horizontal advection, vertical advection, vertical diffusion and dry deposition, aqueous phase chemistry, moist convection and gas phase chemistry. Mass changes in the PBL up to approximately 2.5 km height (model layers 1–7) are shown for total ozone, ozone produced by photochemical reactions and the sum of $O_{3,i}$ and $O_{3,b}$. Negative values in Figure 5 denote a decrease of ozone concentrations as a result of the respective process, positive values an increase. Table II summarises the percentage contributions of $O_{3,pr}$ and $O_{3,i} + O_{3,b}$ to the total ozone mass changes determined from the absolute values of the respective numbers in Figure 5, the sum of both makes up 100%. Ozone production by chemical reactions dominates over the other processes in the PBL. Photochemically produced ozone (Figure 5(b)) is transported to the free troposphere by vertical advection and moist convection, with moist convection transporting about half as much as vertical advection. Horizontal advection exports the same amount of ozone out of the German PBL as vertical advection does. Vertical diffusion together with dry deposition is the most important ozone loss process in the PBL. Cloud chemistry is unimportant during the presented episode.

Differently from $O_{3,pr}$, the concentrations of initial and boundary ozone within the PBL in the area of Germany increase due to horizontal advection, vertical advection and moist convection (Figure 5(c)). The only important loss process of $O_{3,i}$ and $O_{3,b}$ in the PBL is chemical destruction. The loss of $O_{3,i}$ and $O_{3,b}$ due to vertical diffusion and dry deposition appears to be negligible. However, these two

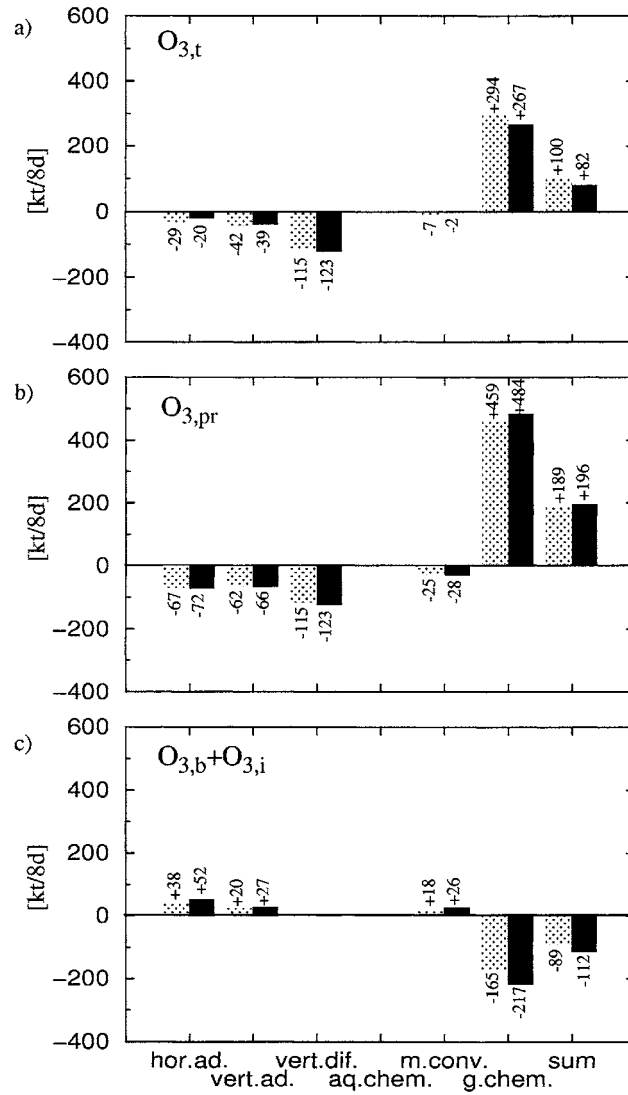


Figure 5. Mass tendencies [kt/8d] over the German subarea due to horizontal advection (hor. ad.), vertical advection (vert. ad.), vertical diffusion and dry deposition (vert. dif.), aqueous phase chemistry (aq. chem.), moist convection (m. conv.), gas phase chemistry (g. chem.) and as sum over all processes of (a) total ozone, (b) photochemically produced ozone and (c) the sum of initial and boundary ozone. Mass changes in the PBL up to approximately 2.5 km are shown as grey shaded bars for the baseline simulation and as black bars for the sensitivity experiment.

processes compensate each other so that the numbers are misleading. Although photochemically produced ozone in the PBL over Germany dominates the changes in total ozone mass (68%, see Table II), the overall effect of $O_{3,i}$ and $O_{3,b}$ on total ozone mass changes represents 32% and is therefore far from being negligible. It is also important to note that the vertical exchange of ozone between the PBL and the free troposphere due to vertical advection, vertical diffusion and moist convection significantly affects ozone concentrations in the PBL.

A sensitivity experiment with an other set of climatological initial and boundary concentrations of ozone has been carried out to further illustrate the influence of the background ozone distribution. Following the recommendations for the EURAD model (Hass, 1991) the vertical summer ozone profiles over Europe shown in Figure 6 have been used for the initialisation and as boundary concentrations. These data have been interpolated linearly in space to the respective grid point locations of the REMO model. Again, the concentrations at the lateral boundary grid points were fixed throughout the simulation. The vertical ozone profiles in Figure 6 present high-, mid- and low latitude summer conditions. The tropopause altitude decreases with latitude. Background tropospheric ozone concentrations at 50° north are assumed to be significantly higher than the tropospheric concentrations at lower and higher latitudes. For comparison, the ozone profile included as initial and boundary profile in the baseline model simulation as discussed above is also shown in Figure 6. Throughout the troposphere the base ozone profile shows values between the respective profiles of the sensitivity experiment. As the model domain (Figure 1) is located with $\pm 15^\circ$ around 50° north, the background concentrations that are specified in the sensitivity experiment are higher than in the baseline simulation. Stratospheric ozone concentrations have been fixed to 100 ppbv in the baseline model case. However, these somewhat unrealistically low concentrations did not influence the tropospheric ozone distribution during the episode under consideration.

Results of the sensitivity study are presented in Figure 5 as black bars and are summarised in Table II. Higher initial and boundary ozone concentrations lead to a higher contribution (36%) to the mass changes of total ozone concentrations compared to the baseline simulation. As discussed before for the baseline simulation, the relevant processes are horizontal advection, vertical advection, moist convection and photochemical destruction. The higher background concentrations increase the amount of photochemically produced ozone, $O_{3,pr}$, present in the PBL. This somewhat surprising result is caused by the methodology of the ozone split and by the various chemical loss processes that are in competition: higher background $O_{3,i}$ and $O_{3,b}$ consume more reactants so that more $O_{3,pr}$ stays in the PBL and more $O_{3,pr}$ is removed from the PBL by horizontal advection, vertical advection, dry deposition and moist convection. Compared to the baseline model simulation the values of the net tendencies of total ozone due to horizontal advection, vertical advection, dry deposition, moist convection and gas phase chemistry are reduced. The influence of different background ozone concentrations on near

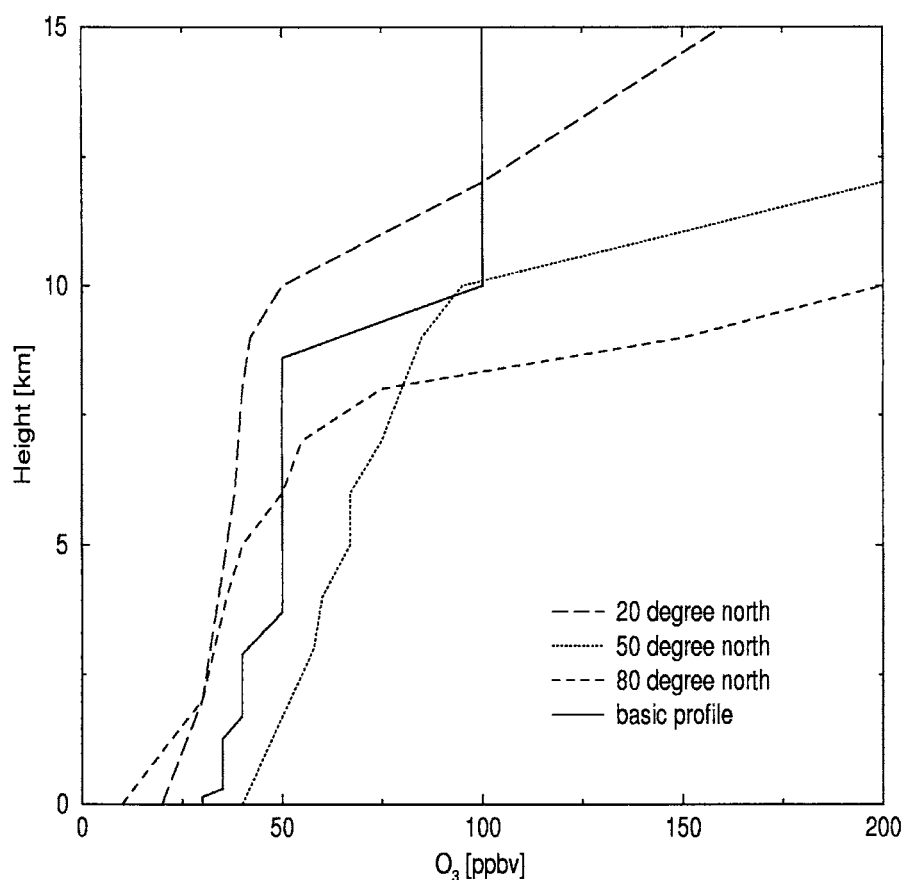


Figure 6. Vertical ozone profiles in ppbv as used for the initialisation and as boundary concentrations for the REMO model. The full line represents the profile included in the baseline model simulation, the broken lines stand for the profiles of the sensitivity experiment.

surface ozone concentrations is presented in Figure 7 as a result of the sensitivity experiment and as difference between the sensitivity and the baseline model run. An 8-day average at 16 UTC is shown (excluding the first 24 hours of the simulations). The maximum near surface ozone concentration in Figure 7(a) of 82 ppbv is relatively small for a summer smog episode because the averaging procedure also includes the low ozone concentration calculated at the beginning of the smog episode (Langmann, 2000). In the German subarea the maximum differences (Figure 7(c)) exceed 4 ppbv corresponding to an average difference of about 5% (Figure 7(b)). Over Great Britain and Ireland a tongue of enhanced western boundary ozone concentration is visible in Figure 7(b). In this area the impact of the modified background conditions reaches more than 15%. However, at the end of July 1994 the photochemical situation in Great Britain was different from that in Germany (Langmann, 2000). While an intensive summer smog period

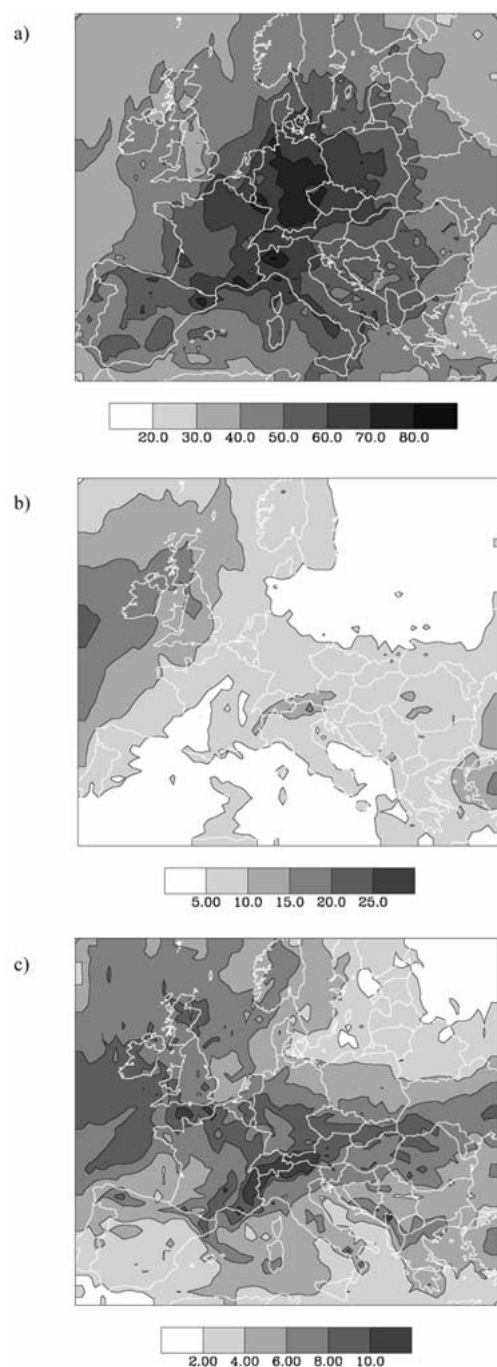


Figure 7. (a) Near surface ozone concentrations in ppbv as determined by the sensitivity experiment as eight days average (21–28.7.1994) at 16 UTC together with (b) the difference: sensitivity run minus baseline model simulation in % and (c) the maximum difference in ppbv at the individual grid points occurring during the eight days at 16 GMT.

was developing over Germany, Great Britain remained under the influence of low pressure with cloud systems. Thus, photochemical activity was reduced and unpolluted maritime air masses from the North Atlantic were advected towards the British Isles. This example demonstrates that under low pressure influence with westerly flow conditions the quality of regional ozone simulations depends even stronger on realistic lateral boundary concentrations of ozone.

4. Conclusions and Outlook

The sensitivity and uncertainty of background ozone concentrations for regional scale air pollution modelling have been analysed for a specific summer smog episode over Europe at the end of July 1994. For this purpose, total ozone was splitted artificially into individual categories. For each category, transport and chemical transformations have been determined separately. The conclusions of Brost (1988) concerning initial and background concentrations have been mainly confirmed. The effects of the initial ozone concentrations decline in importance as the simulation proceeds. However, differently from Brost's (1988) conclusions they remain important until the fifth day of the simulation because of the high surface pressure system over Central Europe during the described summer smog period. Initial and boundary concentrations of ozone dominate total ozone concentrations increasingly with height. However, ozone background concentrations have also a significant influence on total ozone concentrations in the PBL. Background ozone is imported into the PBL by horizontal and vertical advection, vertical diffusion and moist convection and lost there by photochemical destruction and dry deposition. Although background concentrations do not appear in significant amounts in the PBL due to the chosen split approach, they contribute with more than 30% to the total ozone changes in the PBL. If background vertical ozone profiles are modified, near surface ozone concentrations might decrease or increase by more than 5% depending on the weather situation. The higher the background ozone concentrations, the higher is the amount of total ozone and of ozone produced by photochemical reactions in the PBL because photochemical loss processes are in competition. The results demonstrate the importance of the vertical mixing of ozone between the PBL and the free troposphere and emphasise the need of realistic background concentrations of ozone throughout the troposphere to reduce uncertainties in calculated ozone concentrations in the PBL.

For the presented investigation climatological background ozone concentration profiles at the lateral model boundaries have been used. Thus, the temporal variability in ozone concentrations at the lateral model boundaries has been ignored and an even higher uncertainty in near surface ozone concentrations as determined by limited area models can be expected. Another limitation of the current study is the short simulation time of only eight days. A longer lasting simulation, for example the whole summer season, would exclude the impact of the initial ozone

distribution and would allow an investigation of the influence of background ozone concentrations on total ozone under different weather conditions.

The simple formulation of initial and lateral boundary conditions for chemical trace species in regional air pollution modelling is applied due to the lack of detailed information of trace species concentrations throughout the troposphere. Unfortunately, a global observation network including surface based and satellite measurements as it exists for meteorological variables is not yet available for atmospheric trace species. Moreover, such a network will be difficult to develop due to the large number of chemical trace species involved. Assimilation of only near surface measurements as described in Elbern *et al.* (1997) is insufficient. The current study emphasises the need of realistic background concentrations throughout the troposphere to better understand and predict regional distributions of trace species and the major processes involved. One possibility is a global-regional nesting approach as it is also applied for weather forecast simulations. However, the connection between global and regional atmosphere-chemistry models is still missing. Up to now limited area atmosphere-chemistry models have been applied to simulate real weather situations with the purpose of validating and evaluating the calculated distributions during short pollution episodes. Global atmosphere-chemistry models, however, have been applied mainly for climatological simulations. There are already several attempts to force a global model atmosphere to stay close to the real weather situation by applying for example the Newtonian relaxation technique with meteorological analysis data (Jeuken *et al.*, 1996). If global atmosphere-chemistry simulations become available for certain episodes or even better as standard data sets in 6 hour intervals, a one-way nesting technique could be applied: large scale phenomena are simulated by global coarse grid models and the results are used to provide background concentrations for a higher resolution limited area model simulation over the regions of interest.

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